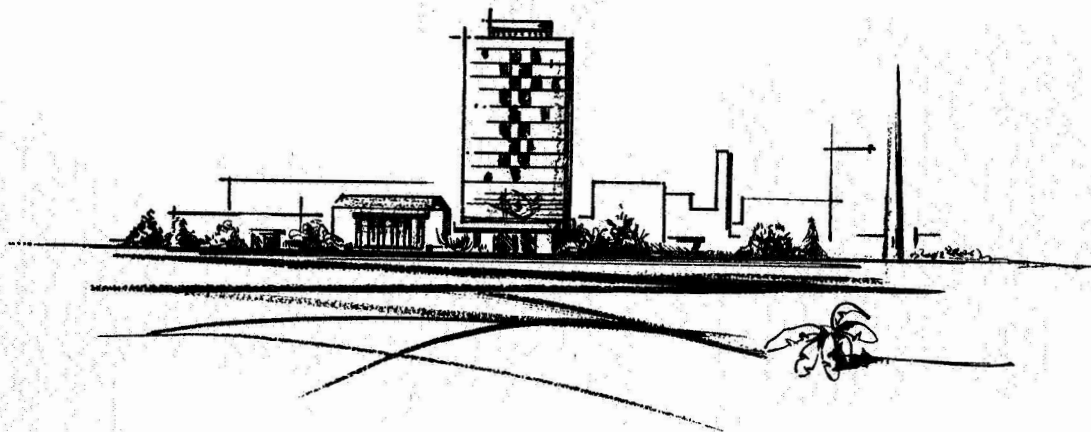# RESEARCH REPORT

ANALYSIS OF RADIATION EFFECTS ON STORED
LIQUID PROPELLANTS ON SPACECRAFT WITH
AUXILIARY RTG OR NUCLEAR POWER SOURCES

to

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

PASADENA OFFICE

May 31, 1968

BATTELLE MEMORIAL INSTITUTE

COLUMBUS LABORATORIES

FINAL REPORT

on

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by

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ABSTRACT

An analysis of radiation effects on liquid propellants of interest as stored propellants on interplanetary unmanned spacecraft has been completed. Radiation damage data for 14 liquid fuels and 15 liquid oxidizers are tabulated. Radiation levels from external space radiation as well as representative α-, β-, γ-radioisotope and nuclear reactor power sources are presented in parametric form. A summary is given of propellant radiation damage as a function of source type and mission. It was concluded that α-heat sources such as ^{238}Pu or ^{244}Cm probably would not require additional shielding to protect the propellants even with large power sources. β-and γ-radioisotope sources and nuclear reactor sources likely will require propellant shielding on long missions.

ANALYSIS OF RADIATION EFFECTS ON STORED
LIQUID PROPELLANTS ON SPACECRAFT
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SOURCES

INTRODUCTION

It is advantageous to use high specific impulse liquid propellants on

long-term space probes using isotope or nuclear power sources for auxiliary power

because of the rigid weight and volume requirements on such missions. However,

more definitive knowledge of environmental effects on the long-term storability of

these propellants is desirable so that design engineers may have reasonable assur-

ance that the stored propellant system chosen for a particular mission will fulfill

its function successfully.

The aim of this study was to ascertain the state-of-the-art regarding

radiation effects data on liquid propellants and to analyze the suitability of using

various propellant systems on space probes with different auxiliary power sources.

Within the scope of this study the following three principal tasks have been performed:

(1) A state-of-the-art survey of existing knowledge of radiation

effects on propellants of interest has been performed and the

results tabulated,

(2) The type and intensities of radiation likely to be encountered from

a variety of isotope and nuclear power sources has been quantitatively

analyzed, and the radiation dose to propellants from external space

radiation has been estimated,

(3) The relationship between the radiation damage effects in the

propellants and the radiation dose levels to be attained during

the various missions has been evaluated. The results are presented in parametric form in all cases where sufficient data are available.

Recommendations are made concerning the suitability of a number of fuel and oxidizer compositions as passively stored propellants on long term space missions. Those cases are noted where experimental data are too sparse to allow a reasonable judgment to be made concerning their suitability. Special note is taken of instances where the evidence indicates that an unusual problem may be present.

SUMMARY OF RADIATION EFFECTS ON LIQUID PROPELLANTS

The primary aim of this phase of the study was to pinpoint the nature and seriousness of potential problem areas which may arise when liquid propellants are stored in a radiation environment. The survey covered a total of 14 fuels and 15 oxidizers (see Tables 1 and 2). Most of these propellants are listed in RFQ No. W067-10(PR67-463). The others were included in one or more of the radiation effects studies reviewed.

An exhaustive literature search was carried out covering all radiation effects data available for all the propellants discussed in this study. Primary literature sources include published literature, government report literature and supplementary information in the REIC files. In order to insure that the information reflects a complete and up-to-date assesment of the state-of-the-art, direct contact was made with major government and industrial installations which have performed research or might have an interest in radiation effects on liquid propellants. A total of about 30 individuals in 15 facilities were contacted in all, during the course of the program.

General Discussion of Radiation Effects
on Propellants

Radiation Energy Deposition

Ionizing radiation transfers its energy to the material traversed in different ways. Fast charged particles transfer their energy predominantly by inducing electronic transition of molecules in or near their path; fast neutrons and γ rays produce charged particles which also behave in this manner. The quantitative dependence of the linear rate of energy transfer* on the nature of the radiation and the material under irradiation are often of interest in radiation effects studies. Table 1 from Ref. 1 gives values of -dE/dx for various kinds of radiation in water to illustrate the strong dependence of this parameter on radiation type. The radiation environment seen by liquid propellants on a space mission may include all of these types of radiation.

TABLE 1. L.E.T. FOR DIFFERENT RADIATIONS[1]

Radiation	L.E.T. in ev/A
0.01 MeV electrons	0.23
0.01 MeV electrons	0.04
1 MeV electrons	0.02
^{60}Co γ-radiation	0.02
1 MeV protons	2.8
10 MeV protons	0.47
1 MeV alphas	26.4
10 MeV alphas	5.6
^{10}B (n,1.5 Mev α)0.85 Mev ^{7}Li	~17

* Linear Energy Transfer (L.E.T.) is the differential rate of loss of energy per unit length of the track of an ionizing particle $\left(\frac{-dE}{dx} \right)$.

Formation of Intermediates and Products

As a result of energy deposition by ionizing radiation excited molecules, ions and electrons are formed. Excited molecules may lose their energy without chemical reaction, decompose to give radicals or stable molecules, or react with other normal or excited molecules. Thus there can be competition between first and second order reactions of excited molecules, which can give rise to dose-rate or L.E.T. effects.

In measuring the chemical effects of radiation, for instance the G-value[*], one is concerned with the overall effect of radiation as it penetrates the substance and its energy is dissipated. Although some understanding of radiation damage mechanisms is essential in understanding and extrapolating experimental data, it is the final chemical products formed by radiation which are of primary interest here.

Radiation Damage in Liquid Propellants

Radiation effects in the propellant systems under consideration may be expected to manifest themselves in one or more of the following ways:

(1) Production of volatile off-gases (leading to pressure buildup),

(2) Radiation polymerization (leading to increase in viscosity or solids deposition),

(3) Radiation synthesis of reactive chemical species (leading to chemical composition changes, possible detonable compounds, or enhanced corrosion effects).

The first of these is by far the most common in the case of fuels, due primarily to their tendency to form hydrogen gas. In the following section, the most critical damage parameter is specified for each propellant in those cases where there are sufficient data to show it.

---

[*] The number of molecules formed or destroyed per 100 ev of energy deposited.

Dose rate and L.E.T. effects may be important for several reasons; some of which are:

(1) There may be different orders of intermediate reaction at different dose rates or L.E.T.'s leading to different products,

(2) Some radiation products may be chemically unstable and spontaneously decompose at a rate fast enough to prevent a significant concentration from building up over a period of time,

(3) Slow post-irradiation chemical reaction involving radiolysis products may produce changes in chemical composition observable only at very low dose rates and long exposures.

Radiation effects testing is ordinarily carried out at dose rates much higher than the propellant will be subject to under conditions of use. Therefore, some consideration should always be given to the type of subtle effects discussed here. If there are reasons to believe that dose rate or L.E.T. effects may be significant for a particular propellant, this possibility should be checked experimentally.

Summary and Evaluation of Radiation Effects Data

Radiation effects data on all the fuels and oxidizers surveyed are presented in Tables 2 and 3 respectively. The significance of the various entries in the Table is as follows:

Columns 1, 2, and 3: The propellants are grouped into chemically related classes and each composition in the class is listed separately. References to radiation effects data on each composition are found in the bibliography.

Column 4: Here, the radiation studies are rated according to relative

completeness of the data obtained. <u>A comprehensive parametric study</u>
is one which covers a range of radiation types, dose rates, temperatures,
etc. <u>A limited parametric study</u> considers perhaps 2 or 3 parameters.
<u>A verification test</u> involves subjecting the sample to one type of radiation
at one dose rate to one dose level. Also, the types of radiation used in
the indicated studies is indicated.

<u>Column 5</u>: The percent decomposition per megarad (Mrad = 10^8 ergs/g) is
used as a convenient standard for comparison and calculation. The values
given here have been derived from direct measurements or they represent an
upper limit estimated from the data which are available.

<u>Columns 6 and 7</u>: The volume of gas formed is given in terms of ml per
megarad in column 6. The major off-gas components are listed in column 7,
where they have been identified, and other products may be formed to a
lesser extent.

<u>Columns 8 and 9</u>: Major liquid radiolysis products are listed in column
8 in cases where they have been identified and other special radiation
effects are denoted in column 9 as observed.

<u>Column 10</u>: The radiation effect(s) judged to present the most serious
engineering problems are given here. The judgment as to a specific pro-
pellant's suitability for a particular mission is based on this "most
critical" damage parameter.

Experimentally Observed Radiation Efffects on Fuels

The summary of radiation damage data for fuels is found in Table 2. The fuels
as a group are much more sensitive to radiation damage than are the oxidizers, and more
of them have been subjected to radiation testing. It was found that there are some data
on all of those listed in Table 2. The effects on each class of fuels are discussed
below where individual differences and trends are pointed out.

TABLE 2. SUMMARY OF RADIATION DAMAGE DATA IN FUELS

Fuel Class	Fuel Composition	Reference Numbers	Radiation Effects on Fuel						Major Radiation Problem
			Type of Radiation Effects Study	Percent Decomposition Per Mrad	Volume of Gas Formed (ml/Mrad)	Composition of Off Gas (8)	Liquid Radiolysis Products Observed(8)	Other Effects Noted	
Hydrocarbon	Methane, CH_4	2 - 6	Comprehensive Parametric, α,x,γ,β	0.010	0.09	H_2	C_2H_6		P-buildup
	Ethane, C_2H_6	2,6	Gas Phase + Verification Test on Liquid, α,γ,x	0.02[1]	0.09[1]	H_2, CH_4	C_2H_4, C_3H_8		P-buildup
	Propane, C_3H_8	2,5	Gas Phase, γ,s	0.02[1]	0.09[1]	H_2,CH_4, C_2H_6	C_3H_6, etc.		P-buildup
	Ethylene, C_2H_4	2,6-9	Comprehensive Parametric, α,β,γ	0.045	0.03	H_2	C_2H_2, etc. Polymer[2]		P-buildup, Polymer form.
Amine	Ammonia, NH_3	10-13	Comprehensive Parametric,γ,x,β	0.002	0.03	H_2, N_2	N_2H_4	(7)	P-buildup
	Hydrazine, N_2H_4	11, 14-16	Limited Parametric,x	0.01[3]	0.10	H_2, N_2	?	(7)	P-buildup
	Monomethylhydrazine, CH_3NHNH_2, MMH	6,15,17,18	Limited Parametric, γ,n,x	0.025[3]	0.28	H_2, N_2, CH_4	None Obs.		P-buildup
	1,1-Dimethylhydrazine, $(CH_3)_2NNH_2$, UDMH	14-17	Limited Parametric, γ	0.04[3]	0.27	H_2, H_2CH_4	None Obs.		P-buildup

TABLE 2. (Cont'd)

Fuel Class	Fuel Composition	Reference Numbers	Type of Radiation Effects Study	Radiation Effects on Fuel					Major Radiation Problem
				Percent Decomposition Per Mrad	Volume of Gas Formed (ml/Mrad)	Composition of Off Gas(8)	Liquid Radiolysis Products Observed(8)	Other Effects Noted	
	Diethylene Triamine $(H_2NCH_2CH_2)_2NH$, DIEN	15-17	Limited Parametric,γ	0.045	0.12	H_2	None Obs.		P-buildup
Amine	Aerozine, 50:50 UDMH:N_2H_4	16-19	Verification Tests, γ,n	.02[4]	0.21[4]	H_2, N_2, CH_4	None Obs.		P-buildup
	Hydyne, 60:40 UDMH:DIEN	15-19	None	.04[5]	0.21[5]	H_2 N_2 CH_4	None Obs.		P-buildup
Boron	Diborane, B_2H_6	6	Verification Test,γ	~.01	≤ 0.1	H_2	None Obs.		P-buildup
Hydride	Pentaborane-9, B_5H_9	16-20	Verification Test + Vapor Phase Studies, β,γ	~.01	(0.3 in gas) ≤ 0.1 in liq.	H_2, B_2H_6	Color[6] Change	(7)	P-buildup
Elemental	Liquid Hydrogen, H_2	21-24	Comprehensive Parametric, n,γ					p-o-H Conversion	Radiation Heating

(1) Based on liquid methane data.

(2) 0.0003 g polymer per gram per Mrad.

(3) Based on off-gas analysis.

(4) Estimated from N_2H_4 + UDMH data.

(5) Estimated from UDMH + DIEN data.

(6) Suspected polymer formation.

(7) Surface effect noted by some investigators, chiefly in vapor phase.

(8) Additional products may be formed to a lesser extent in some cases.

TABLE 3. SUMMARY OF RADIATION DAMAGE DATA FOR OXIDIZERS

Oxidizer Class	Oxidizer Composition	Reference Numbers	Type of Radiation Effects Study	Percent Decomposition Per Mrad	Radiation Effects on Oxidizer				Major Radiation Problem
					Volume of Gas Formed (ml/Mrad)	Composition of Off-Gas (2)	Liquid Radiolysis Products Observed (2)	Other Effects Noted	
Elemental	Liquid Oxygen, O_2	25-27	Comprehensive Parametric, γ,n	0.020		--	O_3	None	Ozone Formation Radiation
	Liquid Flourine, F_2	--	None Noted						Heating
	FLOX (Liq. O_2 + liq. F_2)	--	None Noted						
Flouride	Oxygen Diflouride OF_2	6	Verification Test, γ	<0.1	None obs.	--	None obs.	None	?
	Nitrogen Triflouride NF_3	--	None Noted						
	Tetraflourohydrazine N_2F_4	--	None Noted						
	Chlorine Triflouride ClF_3	6	Verification Test, γ	<0.1	None obs.	--	None obs.	None	?
	Chlorine Tetraflouride ClF_4	--	None Noted						
	Perchloryl Flouride ClO_3F	6	Verification Test, γ	.03	0.065	O_2	None obs.	None	?
	Nitryl Flouride NO_2F	--	None						

TABLE 3. (Cont'd)

Oxidizer Class	Oxidizer Composition	Reference Number	Type of Radiation Effects Study	Percent Decomposition Per Mrad	Volume of Gas Formed (ml/Mrad)	Radiation Effects on Oxidizer			
						Composition of Off-Gas (2)	Liquid Radiolysis Products Observed (2)	Other Effects Noted	Major Radiation Problem
Oxides	Nitrogen Tetroxide,	16,19,28	Comprehensive Parametric, γ,n	<.001	.005	O_2, N_2, N_2O	N_2O_5	--	P-buildup
	MON-10, N_2O_4 + 10%NO		None Noted						
	Hydrogen Peroxide H_2O_2	16,29	Verification Test, γ	.2	Significant	O_2	None obs.	(1)	P-buildup

(1) Thermal decomposition was greater than radiation decomposition.

(2) Additional products may be formed in some cases.

Hydrocarbons. This class of fuels has been more intensively studied than any other. Although no definitive studies of liquid ethane and propane have been performed, the data available from vapor phase studies and work with liquid methane and higher hydrocarbons allow reasonable damage estimates to be made. As a class, the hydrocarbons are quite resistant to radiation decomposition. Ethylene is somewhat unique because it has the greatest tendency to form polymer, and at the same time, yields the least volume of off-gas.

Amines. Although ammonia itself is the only fuel in this class that has been studied extensively, data on gas formation are available on all of the amine fuels. It is obvious that there is a wide range of radiation damage sensitivity in this class of fuels. Ammonia is an order of magnitude more resistant to radiation damage than the methyl hydrazines, with hydrazine falling between these extremes. As noted in the table, some radiation-surface effects have been observed in the case of vapor phase irradiation of ammonia and hydrazine. This type of reaction could conceivably cause problems in case these fuels were stored for extended times with a high ullage in a radiation environment.

Boron Hydrides. There have been no comprehensive liquid-phase radiation effects studies on these fuels. The proof tests indicate that they are more resistant to damage than the methyl amines, however, and perhaps are comparable in radiation resistanc to the hydrocarbons. Neutron irradiation deserves a special consideration in this case. Boron is a good slow neutron absorber undergoing the $^{10}B(n,\alpha)^{7}Li$ reaction. The energy released in this reaction is substantial (1.50 mev α and 0.85 Mev ^{7}Li recoil), and both the α-particle and the ^{7}Li recoil are high L.E.T. species depositing a large amount of energy in a short distance (see Table 1). There have been no studies of L.E.T. effects in the boron hydrides so it is not known whether this behavior could cause special problems.

Liquid Hydrogen. There can be no off-gas or complex molecule formation in liquid hydrogen. The possibility of H-atom diffusion into tank walls has been considered.[23] A radiation induced transition from para to ortho hydrogen has been observed. However, this is not of significance for long space missions because the ortho hydrogen decomposes spontaneously back to the para form in a relatively short time.[24] Thus, the primary concern here is probably from radiation heating, which can be treated simply as a heat transfer problem.

Experimentally Observed Radiation Effects on Oxidizers

It was previously pointed out that liquid oxidizers are in general less sensitive to radiation damage than are liquid fuels. However, this may not be true for all fuel-oxidizer combinations. In addition, the lack of definitive radiation effects data for several of the oxidizers listed in Table 3 makes specific comparisons difficult. The information that is available is discussed below.

Elemental Oxidizers. Off-gassing cannot occur in any of the elemental oxidizers, so pressure buildup in them should not be a problem. At higher dose rates, there might conceivably be some attack on tank surfaces by short-lived ions or atoms produced by radiation. In the case of liquid flourine, it is unlikely that any stable molecular species could form, so that simple radiation heating will probably be the only effect of concern in the bulk oxidizer.

The radiation chemistry of liquid oxygen has been studied quite extensively. As indicated in Table 3, the major overall radiation effect is the conversion of oxygen to ozone. Although study has shown that a substantial buildup of ozone can occur in irradiated liquid oxygen, there are several reasons why this will probably not effect

its performance appreciably. In the first place ozone is quite soluble in liquid oxygen,[30] so that it will tend to remain dispersed in solution. Secondly, ozone itself is sensitive to radiation decomposition,[31] so that its concentration cannot build up indefinitely. Finally, tank materials can be chosen which are immune to ozone attack.

In FLOX there might be a small amount of OF_2 produced by radiation. However, this would not be expected to cause any problems because it is probably soluble in the propellant.

Fluoride Oxidizers. Verification Tests have been conducted on liquid OF_2, ClF_3, and ClO_3F. No information was found on NF_3, N_2F_4, NO_2F, and ClF_5. As a class these materials are apparently quite radiation resistant, with perchloryl fluoride being probably the least resistant of those tested. It would certainly be desirable to have definitive experimental data for this group because it includes some oxidizers of great interest to the space program.

Oxides. Sufficient data have been obtained in this case to prodict the nature and extent of radiation damage with considerable confidence. Nitrogen tetroxide appears to have good radiation resistance. MON-10 would be expected to behave about the same; however, no reference was found to any test of this composition. Hydrogen peroxide, which is of doubtful interest as a stored propellant anyway, suffered much more radiation decomposition than did N_2O_4.

ANALYSIS OF RADIATION ENVIRONMENT ON BOARD SPACE PROBES WITH AUXILIARY NUCLEAR POWER SOURCES

To assess the relative merit of candidate propellants for use in deep space missions on the basis of radiation damage and/or threshold, the type as well as intensity of radiation which the propellant will encounter during the lifetime of the mission must be accurately defined. In addition to various kinds of space radiation the propellants to be utilized in nuclear-powered space probes will be subjected to a continuous flux of radiation which originates from the nuclear power source. Both on-board and external sources of radiation are treated in this section.

Probable Types and Intensities of Radiation from Nuclear Power Sources to be Utilized in Deep Space Missions

Present design plans include the use of two distinct types of nuclear power sources depending on the power level, lifetime, and other specific requirements of the space mission. The two types of power sources referred to are long-lived radioisotopes and nuclear reactors of the SNAP variety. Characteristic radiation emanating from these sources can vary widely with respect to type, intensity, and spectra. It is, therefore, necessary to distinguish between the various types of radiation before a comparative evaluation of their radiation dose rate and corresponding radiation damage to propellants is made.

Primary effort in this area was directed toward accurately defining maximum dose levels to be expected from various radioisotope fuel materials which are candidates for radioisotope power systems and from a typical nuclear reactor power source. At least one representative of each type of radioisotope fuel has been included in this analysis. Selected candidate fuels include the following:

(a) Oxides of ^{238}Pu and ^{244}Cm; radioisotopes which decay by emission of α-particles, i.e., high energy He nuclei. The α-particles will be completely absorbed within a short distance in the fuel, but some high energy neutrons will be produced by nuclear interactions of the α-particles. These (α,n) neutrons represent the major source of penetrating external radiation of α-sources.

(b) ^{90}Sr, in the form of $SrTiO_3$, decays by emission of β-particles or high energy electrons. Although more penetrating than α-particles, β-radiation also will be essentially all absorbed in the fuel. However, when β-particles are slowed down by interaction with atomic nuclei, electromagnetic radiation called bremmstrahlung is emitted. This "braking radiation" represents the major source of external penetrating radiation for this fuel.

(c) ^{60}Co, in the metallic form, decays by emission of β-and γ-radiation. The latter is penetrating electromagnetic radiation. Hence, the external radiation field arises as a result of the decay process itself.

The radioisotope ^{238}Pu, which is to be used in the SNAP-27, has received particular attention although the other candidate radioisotope fuels have also been considered as potential auxiliary space power sources. Some of the characteristics of the four radioisotopes considered for use in space power sources are given in Table 4.

An adequate description of a radiation source includes the types of radiation
emitted as well as the intensities and spectra of the emitted radiation. In addition,
all significant impurities, whether decaying or increasing with time, have been in-
cluded and the effect that these impurities have on the overall spectrum and intensity
of the major isotopes are recorded.

TABLE 4. CHARACTERISTICS OF RADIOISOTOPES USED
IN HEAT SOURCES

Isotope	^{60}Co	^{90}Sr	^{238}Pu	^{244}Cm
Half-Life, Years	5.3	27.7	86	18
Compound Form	Metal	$SrTiO_3$	PuO_2	Cm_2O_3
Type of Radiation*	γ, β	β, x	α, n**	γ, α, n**

* α Alpha, β Beta, γ Gamma, n neutron, x Bremmstrahlung

** From (α,n) reaction

Bases of Radiation Dose Estimates

During this study the radiation types, spectra, intensities and integrated
doses for radioisotopes and nuclear reactors were determined on the basis of a source
power level of one thermal watt and source to dose-point separation distance of one
foot. In making the estimates given below the radioisotope fuel has been considered
as an unshielded point source of radiation. This assumption allows a straightforward
calculation to be made of radiation   levels as a function of source power and separation
distance. However, since self-shielding by the source material itself is disregarded,
dose rate estimates will always be somewhat higher than would be true for a real source.
This, in effect, provides a worst case estimate; that is, an estimate of the highest dose

which could be delivered from a given radioisotope fuel. Dose rates expected from a nuclear reactor source are based on data derived from real systems, i.e., they take into account self-shielding, but not external shielding. The effect of self-shielding and source geometry are discussed in a later part of this section of the report.

Radiation doses are expressed in RAD per thermal watt at a distance of one foot from the one watt source. These dose rates were determined by integrating the following function:

$$\gamma \text{ Dose Rate} = \int_0^\infty S(E)_\gamma \cdot D(E) dE \qquad (1)$$

where: $S(E)_\gamma$ = Radiation flux-to-dose rate conversion function for γ-radiation (Ref. 32)

   $D(E)$ = Radiation flux as a function of energy (see, for example, Figures 2 or 3)

In the case of neutron sources, dose rates are commonly given in REM, a biological dose unit. REM Dose is equal to the RAD dose times the Relative Biological Effectiveness, or RBE factor. In general the RBE varies with energy. Therefore, to convert integrated neutron doses in REM to RAD doses, one must divide by the energy dependent RBE factor.

$$\text{Neutron Dose Rate} = \int_0^\infty \frac{S(E) \cdot D(E) dE}{RBE(E)} \qquad (2)$$

where: $S(E)_n$ = Neutron flux-to-REM dose rate conversion function (Ref. 32)

   $D(E)$ = Neutron Flux as a function of energy (see, for example, Figure 1)

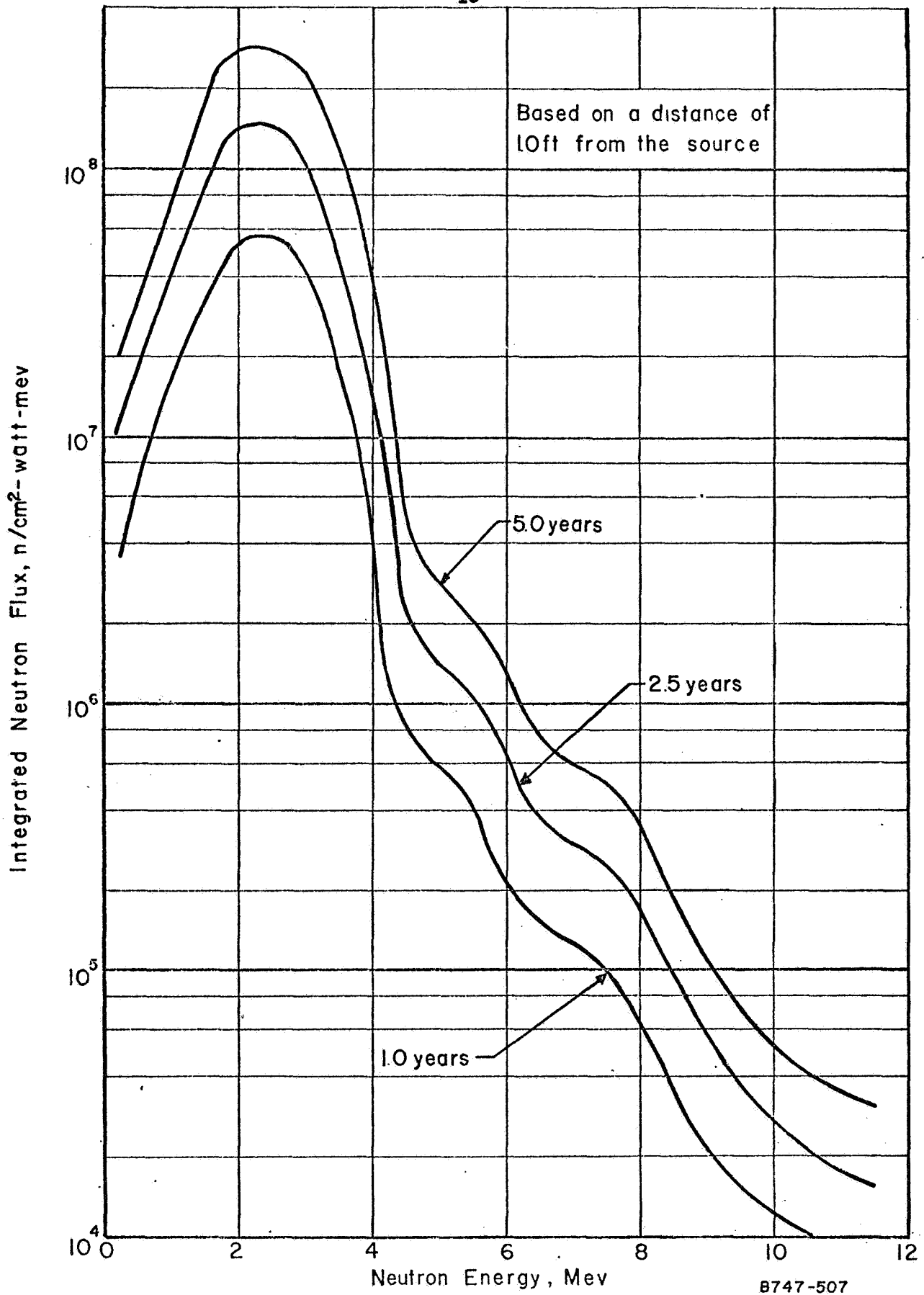 $RBE(E)$ = Energy dependent RBE factor (Ref. 32)
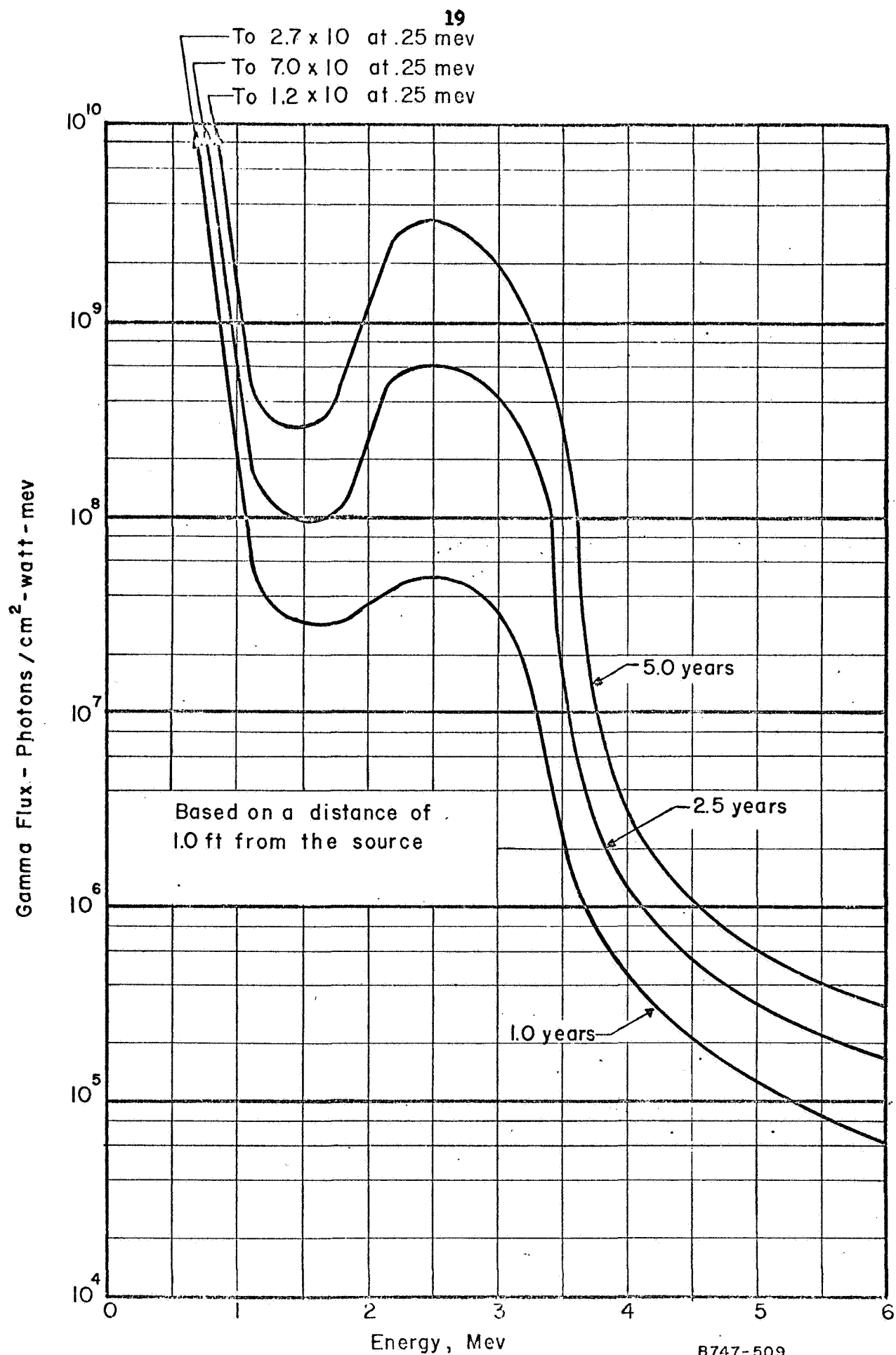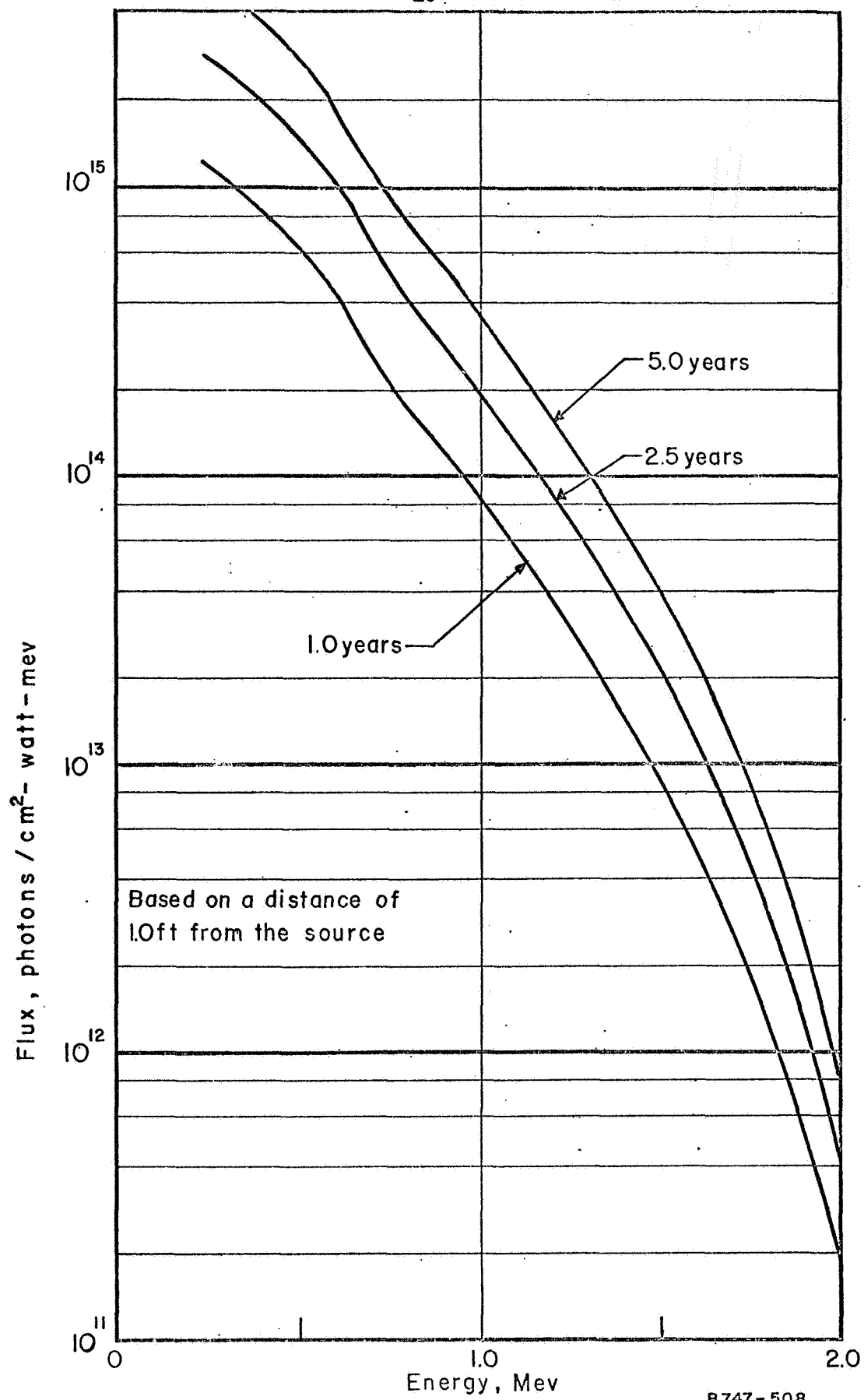
FIGURE I PuO₂ - NEUTRON RADIATION

FIGURE 2 PuO₂ - GAMMA RADIATION

B747-509

FIGURE 3. Sr TiO₂- BREMMSTRAHLUNG RADIATION

Analysis of Individual Candidate Power Sources

$^{238}PuO_2$. (33) The integrated neutron and gamma fluxes of the plutonium oxide

fuel are shown in Figures 1 and 2 for periods of 1, 2.5, and 5 years. In the case

of ^{238}Pu, a trace impurity, ^{236}Pu, forms ^{208}Tl (a gamma emitter) through a long de-

cay chain. The increasing contribution from the buildup of ^{208}Tl 2.6 Mev γ-activity

is easily seen in Figure 2. The absorbed doses from neutron and gamma radiation were

determined from the spectra of Figures 1 and 2 by the procedure described above and

are presented in Table 5.

Cm_2O_3. (34) The neutron and gamma spectra for Cm_2O_3 have been determined but

will not be included in this report. However, unclassified dose rate information appears

in Table 5 for each of the time intervals studied.

$^{90}SrTiO_3$. (35) The bremmstrahlung flux was calculated for the absorption of the

decay beta particles from strontium-yttrium-90 in the strontium titanate ($SrTiO_3$) source.

Bremmstrahlung spectra determined for periods of 1.0, 2.5, and 5.0 years are shown in

Figure 3. The latter spectra were used to determine the integrated absorbed dose for

the three time intervals (see Table 5). This dose was calculated assuming that all

beta particles are absorbed within the heat source. This assumption leads to somewhat

high dose rate estimates for larger Sr sources since the high Z of the heat source yields

a higher energy bremmstrahlung spectra than the generally lower Z supporting structures.

Another strontium isotope, ^{89}Sr, is always present initially in ^{90}Sr fuel, and

its contribution o the dose rate has been included. However, contribution from the

^{89}Sr diminishes with time since the isotope has a 51-day half life compared with a half

life of 28 years for ^{90}Sr.

^{60}Co (Metal).[35]   Cobalt-60 decays by emission of a β-particle, followed by a 1.17 and a 1.33 MeV gamma ray from each ^{60}Co atom. The low energy β-rays and bremmstrahlung do not contribute significantly to the external dose rate. The total integrated flux for each of the gamma rays will, of course, be the same and are given below as a function of time. These flux data were used to determine the integrated absorbed doses given in Table 5.

Energy-Mev	flux, Photons/cm^2-Watt at 1 ft.		
	1.0 yr.	2.5 yr.	5.0 yr
1.17	6.04×10^{15}	1.37×10^{16}	2.39×10^{16}
1.33	6.04×10^{15}	1.37×10^{16}	2.39×10^{16}

Nuclear Reactor.[36]   Probable doses from a reactor power source have been determined based on the SNAP-8 reactor. The radiation spectrum and doses from this reactor are typical, at a given power level, of those expected from other SNAP reactors such as the SNAP-2, 10, or 50. The integrated gamma and neutron fluxes for 1.0, 2.5, and 5.0 year missions are shown in Figures 4 and 5. For the purpose of comparison, Table 5 lists the integrated reactor doses along with the radiation dose data for isotope sources.

TABLE 5.   INTEGRATED DOSES FROM VARIOUS SOURCES

Source	Radiation	Dose (Rads/Watt at 1 foot)		
		1.0 Year	2.5 Year	5.0 Year
$^{238}PuO_2$	Neutron	6.0	15	30
	Gamma	0.37	0.91	1.8
	(Total)	6.4	16	32
$^{244}Cm_2O_3$	Neutron	32	79	1.6×10^2
	Gamma	23	57	1.2×10^2
	(Total)	55	1.4×10^2	2.8×10^2
$^{90}SrTiO_3$	Bremmstrahlung	1.5×10^5	3.8×10^5	7.5×10^5
^{60}Co (Metal)	Gamma	7.3×10^6	1.6×10^7	2.8×10^7
Reactor	Neutron	2.9×10^6	7.4×10^6	1.5×10^7
	Gamma	5.9×10^6	1.5×10^7	2.9×10^7
	(Total)	8.8×10^6	2.2×10^7	4.4×10^7

Application of Radiation Dose Data to Postulated Power Source

The dose rates listed in Table 5 may be extrapolated to dose rates for higher thermal power sources at various distances by multiplying by the ratio:

$$\frac{\text{Power (in watts)}}{\text{Distance}^2 \text{ (in feet)}} \cdot$$

For example, the total maximum dose from a typical SNAP-27 source containing the equivalent of a kilowatt of $^{238}PuO_2$ radioisotope fuel, can be found by multiplying the sum of the neutron and gamma contributions by the power level in watts. The result
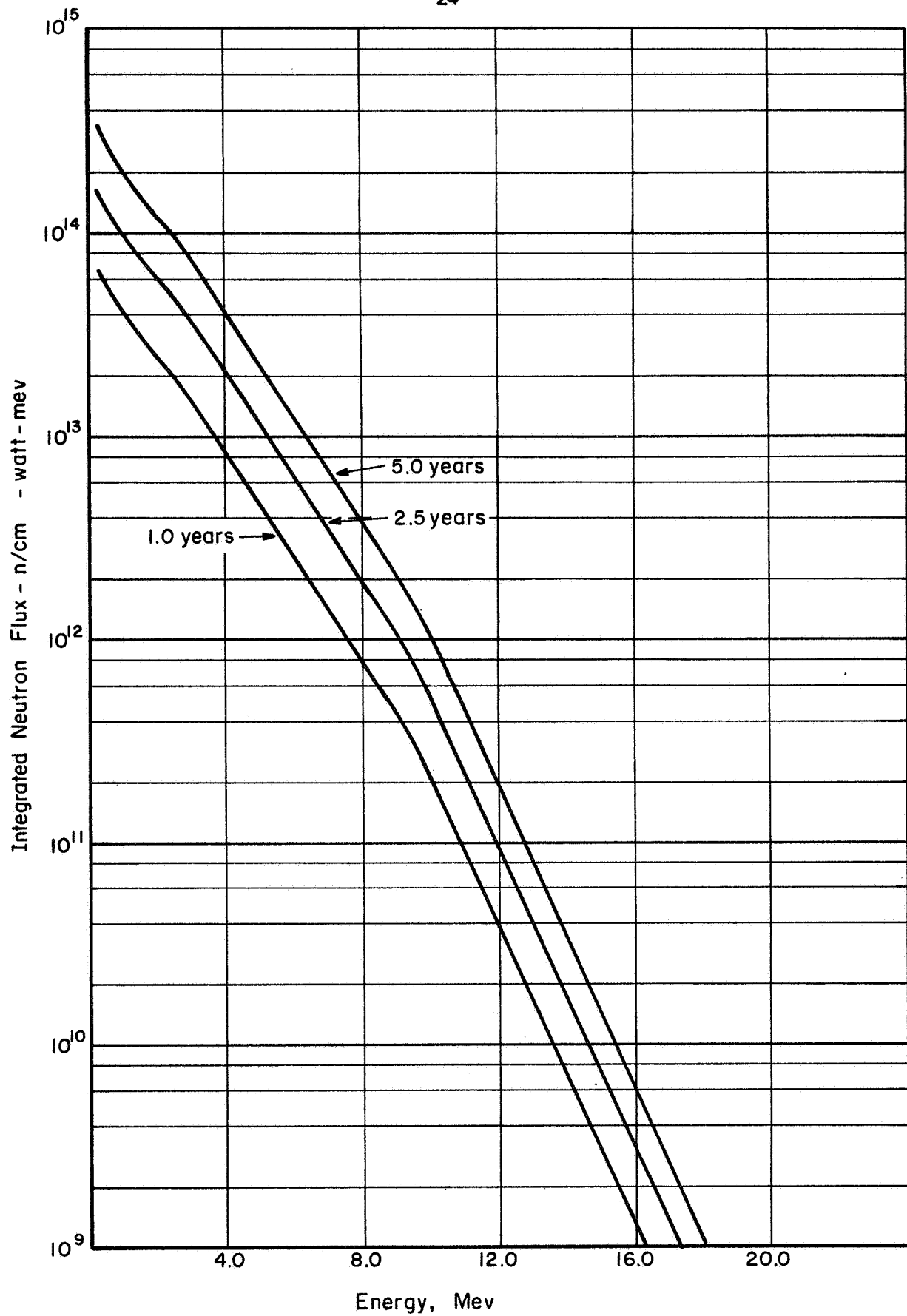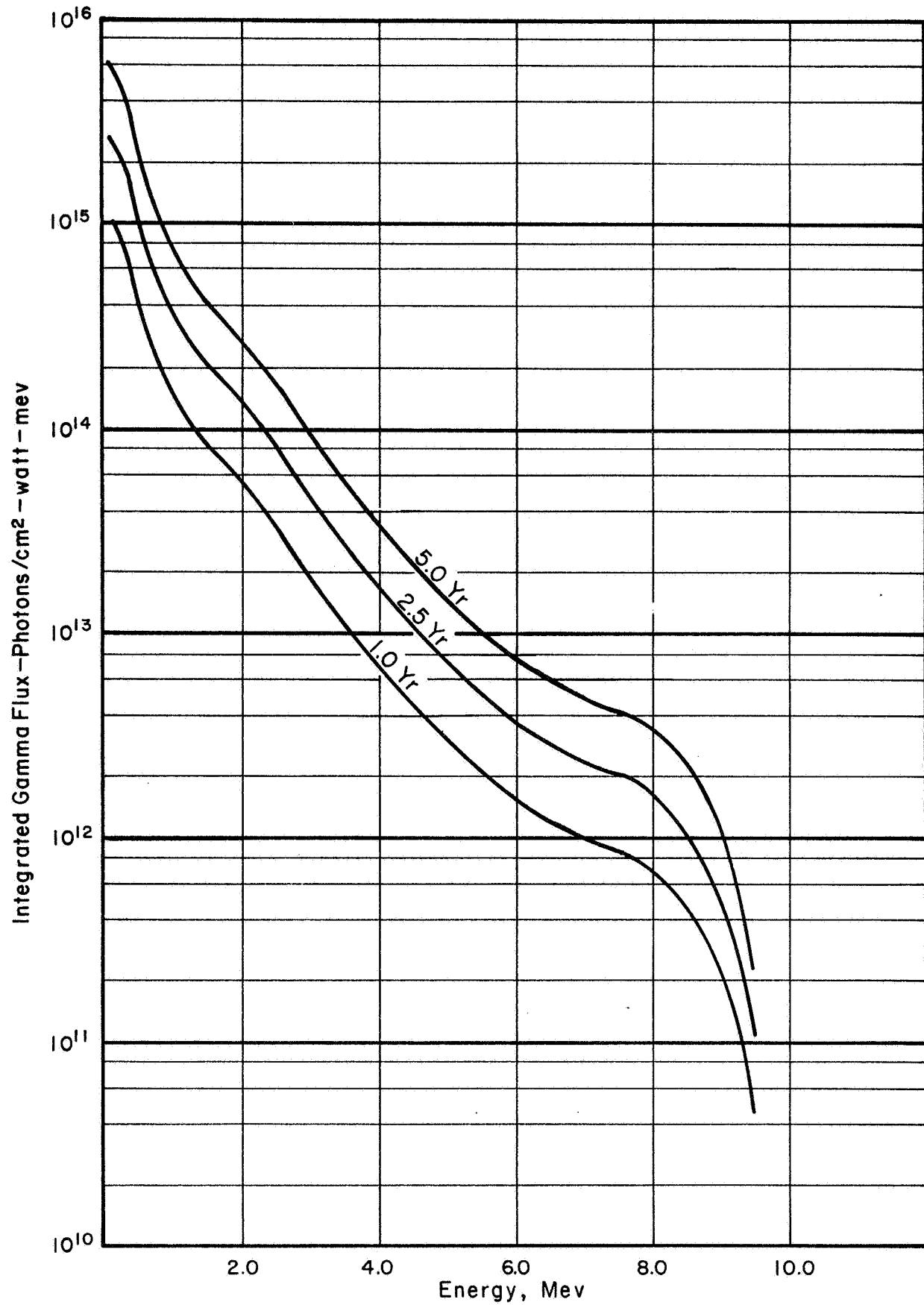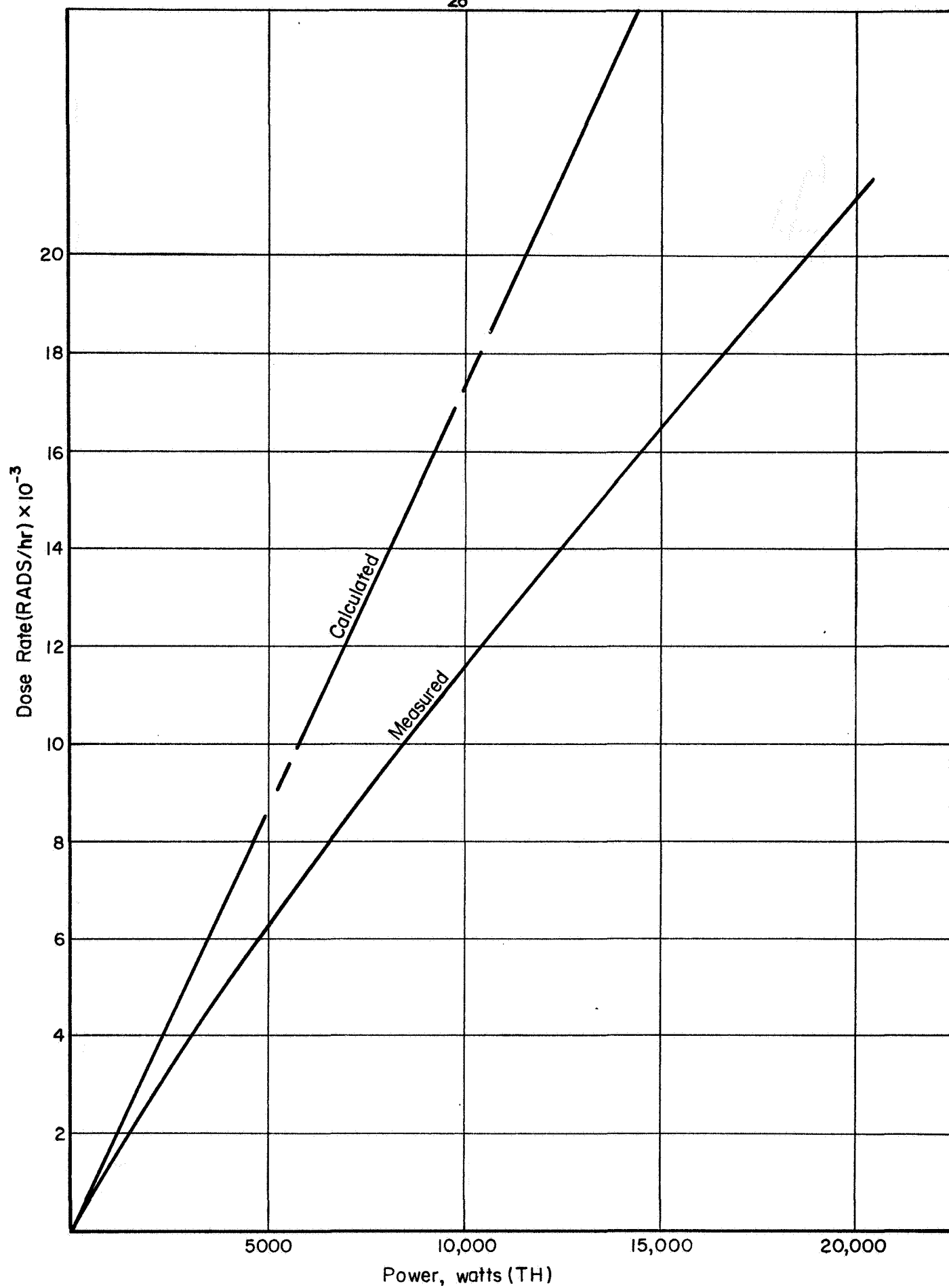
FIGURE· 4. SNAP-8 NEUTRON FLUX

FIGURE 5. SNAP-8 GAMMA FLUX

FIGURE 6. DOSE AS A FUNCTION OF POWER (WATTS) MEASURED AT ONE FOOT FROM CENTER OF SrTiO₃ SOURCE

will be the dose integrated over the chosen time span. Thus, for the kilowatt source and a 2.5 year mission, the integrated dose at one foot would be estimated as 1.6×10^4 rads. The estimated dose at any other distance can be found by the inverse square of the distance relationship.

It was emphasized above that these data, with the exception of the reactor data, are computed on the basis of point sources of radiation and, therefore, do not account for self-shielding within the source material. Neglect of self-shielding in the source ordinarily results in a calculated radiation dose rate which is too high. This result is shown graphically in Figure 6 where the dose rate from a $^{90}SrTiO_3$ source is plotted as a function of power level for both the unshielded (i.e., point source) and self-shielded cases.[37] At a thermal power level of 20 kw*, the point source assumption is seen to result in an overestimate of the radiation dose rate by a factor of 1.6.

The data for ^{90}Sr are probably typical, and illustrate the fact that the dose rate estimates given in Table 5 for the various radioisotope power sources are conservative.

The geometry of a particular power source is another effect not specifically considered in this screening study. However, differences in source configuration can significantly effect external radiation levels around an actual radiation heat source. In addition, since real power sources are not, in general, perfectly spherical in shape, different orientations of the power source with respect to a particular absorber will lead to different radiation dose rates at the same distance from the source center. More refined determination of the radiation environment around a real power source should consider all significant perturbations to the radiation field. It should be clear from this brief discussion that accurate assesment of radiation dose rates from a potential auxiliary power source requires knowledge of the configuration of the spacecraft as well as that of the power source.

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* This value is equivalent to 1 kw(e) if thermoelectric conversion is assumed at 5 percent efficiency.

Estimate of Space Radiation Dose to Propellants

The penetrating radiation environment in space consists of three major components: galactic cosmic rays, solar cosmic rays, and the so-called Van Allen entrapped radiation.[38, 39] However, the intensity of galactic cosmic rays is small ($\sim$2 particles/cm^{2}sec) and the estimated dose rate in free space is about 45 mR per day.[38] The Van Allen radiation is found only in the immediate vicinity of the earth or, in general, around a planet having a substantial magnetic field. Thus, the solar cosmic rays are the major source of penetrating radiation outside the earth's magnetosphere.

Solar cosmic rays are high-energy particles emitted when disturbances of poorly understood origin -- solar flare events -- take place on the sun. The particle flux is composed of protons, a varying number of alpha particles and a small admixture of heavier nuclei; however, for the purpose of computing dose, often only protons are considered.

All aspects of the solar proton problem have received intensive study during the last few years because of the potential hazard this radiation presents to manned space flight. As a result, fair estimates can be made of the dose to be expected from this source during extended interplanetary space missions. The dose estimates presented here are based on The Lewis Proton Shielding Code (LSPC)[40] as applied by Scott and Allsmiller.[41-43]

Because the intensity and energy spectrum of solar protons vary markedly from event to event, a statistical analysis of the solar proton events for the purposes of making dose estimates was carried out by Modisette and co-workers.[44] In this model, which is used by Scott,[43] the integral proton spectrum may be represented as

$$J_p(> E) = J_o \exp \lceil -P(E)/P_o \rceil \quad , \quad\quad\quad\quad (3)$$

where:

$J_p(>E)$ = number of protons per unit area in the flare having kinetic energy greater than E,

$P(E)$ = rigidity,

$= pc/Ze$,

p = particle momentum,

c = speed of light,

Z = charge number (1 for protons),

e = electronic charge

J_o, P_o = parameters which characterize a particular flare.

Modisette el al.[44] give values of the parameters J_o and P_o for a large number of flares. They find that P_o varies between 50 and 200 MV and that the total number of protons with energy greater than 30 MeV varies from 10^6 protons/cm^2 to 10^9 protons/cm^2.

Fortunately, the solar flare proton flux falls off rapidly with increasing energy, and thus a modest amount of shielding drastically reduces the solar proton dose. This is illustrated by the data in Table 6 from Ref. 43.

TABLE 6. PROTON DOSE FROM SOLAR PROTONS OF DIFFERENT MAGNETIC RIGIDITIES

Shield Thickness** g/cm^2 Al	Dose (rad per Flare)*	
	P_o = 50 MV	P_o = 195 MV
0	3620	479
5	24.6	86.3
10	4.0	47.9
20	0.53	23.5
50	0.12	7.0
100	0.06	2.1

* 10^9 p/cm^2/Flare
** g/cm^2 (Al) x 0.146 = inches (Al)

As a "worst case" approximation, assume that there are five major solar flares a year; each containing protons with the maximum magnetic rigidity and intensity of Modisette's model. It can be seen from Table 6 that the total dose per year behind 5 gm/cm^2 Al shielding (e.g., equal to 1/8" Al meteroid bumper + 1/8" Al tank wall + 2" B_2H_6) would be <500 rads. From an engineering standpoint, the dose delivered to a stored liquid propellant by solar protons will not present a significant problem since no more than about 10 major solar flares per year of all types are expected even during a period of solar maximum.[38]

SUMMARY OF PROPELLANT RADIATION DAMAGE AS A FUNCTION OF AUXILIARY POWER SOURCE TYPE AND MISSION

Non-Condensable Gases Formed during Propellant Radiolysis

In general, the most significant radiation damage problem from an engineering standpoint is probably the pressure buildup to be expected from radiation formation of non-condensable gases. The major components of radiation-produced off-gas from each fuel are listed in Table 2. Hydrogen gas will be always present because all the fuels contain hydrogen. In addition, nitrogen is commonly formed from irradiation of amines, and methane may be given off if the fuel contains carbon.

The relative sensitivity of various propellants toward radiolytic gas formation is shown in Figure 7. These are the compositions where sufficient data are available to make a reasonable estimate of this property in those cases where it may be a problem. It was previously pointed out that the elemental liquid propellants; i.e., H_2, F_2, O_2, would not be expected to form non-condensable gases under irradiation.

An estimate is made in Table 7 of the expected amount of non-condensable gas formed during a year's mission using 3 different radioisotope power sources at specific power levels. The radiation doses for a $^{238}PuO_2$, a $^{244}Cm_2O_3$ and a $^{90}SrTiO_3$
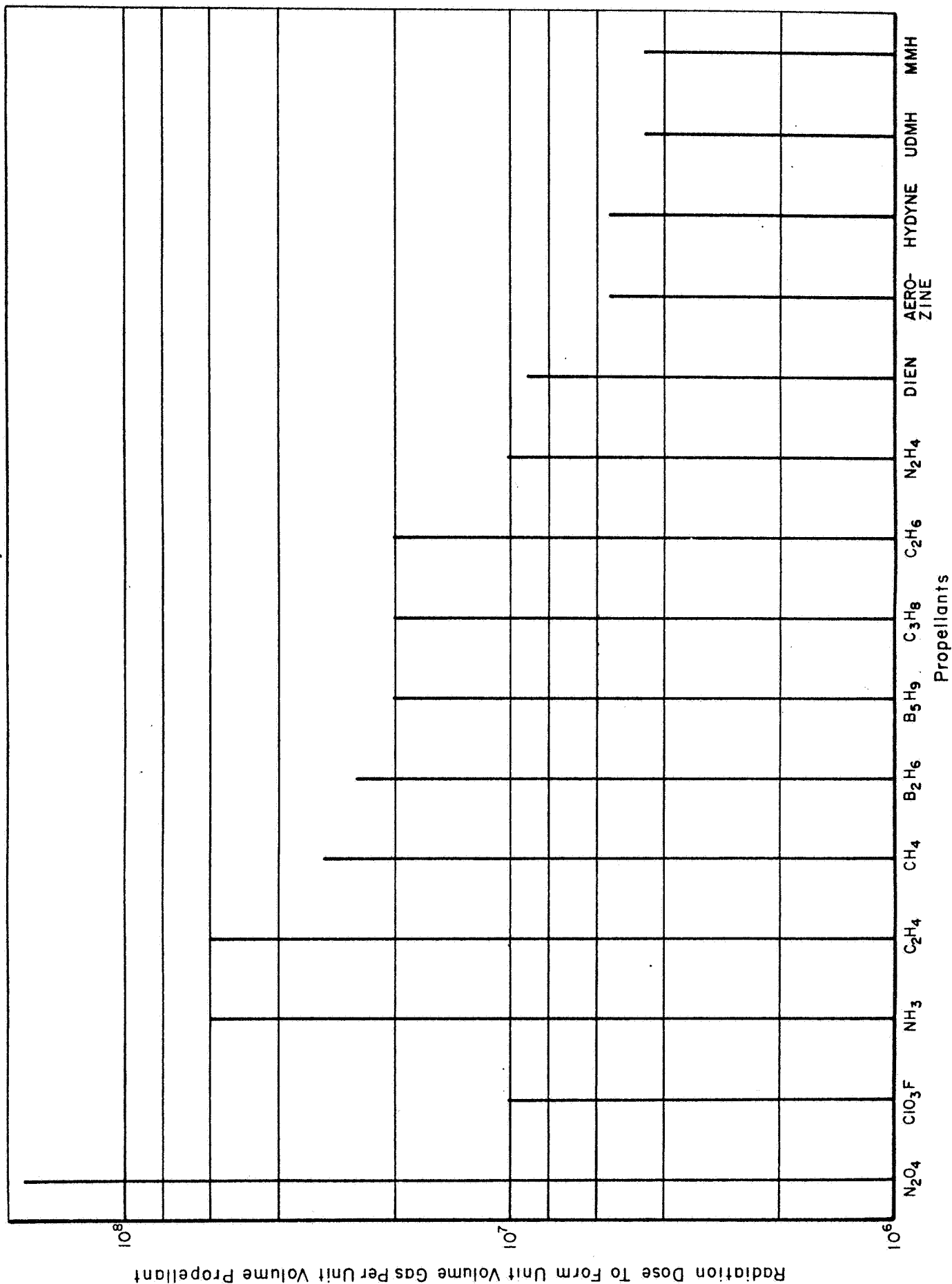
FIGURE 7. RELATIVE RADIATION DOSE REQUIRED TO PRODUCE A UNIT VOLUME OF NONCONDENSABLE GAS PER UNIT VOLUME OF PROPELLANT

TABLE 7. ESTIMATED VOLUME OF OFF-GAS FORMED IN VARIOUS PROPELLANTS BY ^{238}Pu, ^{244}Cm, AND ^{90}Sr HEAT SOURCES

Propellant	Estimated Unit Volume of Off-Gas Per Unit Volume of Liquid Per Year		
	$^{238}PuO_2$ (1)	$^{244}Cm_2O_3$ (1)	$^{90}SrTiO_3$ (2)
Methane, CH_4	$<.01$	~0.05	1.0
Ethane C_2H_6	$<.01$	~0.06	1.5
Propane, C_3H_8	$<.01$	~0.06	1.5
Ethylene, C_2H_4	$<.005$ (3)	~0.02 (4)	0.5 (5)
Ammonia, NH_3	$<.005$	~0.02	0.5
Hydrazine, N_2H_4	$<.02$	~0.1	3.0
MMH, CH_3NHNH_2	$<.03$	~0.25	6.7
UDMH $(CH_3)_2NNH_2$	$<.03$	~0.25	6.7
DIEN $(H_2NCH_2CH_2)_2NH$	$<.02$	~0.1	3.3
Aerozine	$<.03$	~0.2	5.5
Hydyne	$<.03$	~0.2	5.5
Diborane, B_2H_6 (6)	$<.02$	~0.1	1.2
Pentaborane, B_5H_9 (6)	$<.02$	~0.1	1.5
N_2O_4	$<.001$	<0.01	~0.1
ClO_3F	$<.02$	~0.1	3.0

(1) 200,000 watt (th) source at 1 foot (Equivalent to 10 KW(e) at 5 percent conversion efficiency) Dose: $^{238}PuO_2$ - 1.3 x 10^5 rad; $^{244}Cm_2O_3$ - 1.1 x 10^6 rad.

(2) 2,000 watt (th) source at 1 meter (3.25 feet) Dose: $^{90}SrTiO_3$ - 3 x 10^7 rad.

(3) An estimated 5 x 10^{-5} g polymer also formed per gram fuel.

(4) An estimated 4 x 10^{-4} g polymer also formed per gram fuel.

(5) An Estimated .01 g polymer also formed per gram fuel.

(6) Doses in Table 5 from ^{238}Pu and ^{244}Cm have been multiplied by 2 to account for energy deposited in these fuels by the $^{10}B(n,\alpha)^7Li$ reaction. (See Table 1)

source are estimated from the data given in Table 5, assuming no self-shielding or external shielding. Since self-shielding will be significant in all of these sources at the power levels specified, (see for example, Figure 6), the estimates would be expected to be too high. It can be concluded that the α-source should not cause significant problems even at power levels corresponding to 10 KW(e) for 5 year missions. In the case of the ^{90}Sr β-source, the estimates in Table 7 indicate that power sources in the range of 100 watts (e) might present problems for the more radiation-sensitive fuels. Larger ^{90}Sr sources would probably require some external radiation shielding. It is not feasible to make estimates on the use of unshielded ^{60}Co or nuclear reactor sources because their radiation output is too high. Projected applications of such power sources will certainly require detailed consideration of shielding and spacecraft configuration.

Radiation Heating of Propellants

Most of the energy deposited in any material by radiation will eventually appear as heat. At low dose rates the amount of heat involved is so small that it can usually be neglected. However, at high radiation dose rates, radiation heating may present significant design problems. In the case of the propellants listed in Table 7, chemical changes caused by radiation will lead to significant degradation of performance long before heating effects themselves would become important. With elemental fuels and oxidizers such as H_2 or F_2, on the other hand, it is possible that radiation heating may be the most serious engineering problem. However, as mentioned before, radiation heating will only be significant at relatively high dose rates, such as might be encountered from nuclear reactor power sources. Typical heating rates for various dose rates are shown below.

TABLE 8. ESTIMATED HEAT DEPOSITION RATES IN FUELS
FROM VARIOUS RADIATION HEAT SOURCES

| | Radiation Heat Sources | | | | |
	$^{238}PuO_2$	$^{244}Cm_2O_3$	^{90}Sr	^{60}Co	Reactor
Dose/Year, (Rad)[a]	6.4	55	1.3×10^5	7.3×10^6	8.8×10^6
Heat Input:[b] $\frac{cal/g\text{-}hr}{Watt\ at\ 1\ foot}$	5.7×10^{-14}	3.8×10^{-13}	4.4×10^{-10}	2.9×10^{-8}	5.0×10^{-8}

(a) From Table 5.

(b) To convert to $\frac{Btu/lb\text{-}hr}{Watt\ at\ 1\ foot}$ multiply by 1.5×10^2.

Table 8 summarizes the approximate heat deposition rates to be expected in liquid fuels from the various nuclear heat sources discussed in the preceding section of this report. These data are taken from Reference 45 which gives heat deposition rates in various materials from neutron-and γ-irradiation. The neutron values are based on the heating rates in polyethylene whose elemental composition is similar to that of hydrogen containing fuels. γ-heating rates are relatively insensitive to variations in composition.

Miscellaneous Damage Effects

Radiation Polymerization

Ethylene is the only fuel where polymerization is expected to be a problem. Vapor phase radiation polymerization of ethylene has been extensively studied and has been found to proceed very rapidly under some conditions. In contrast, liquid phase polymerization is very slow as indicated in Table 2. However, even a relatively small amount of a high polymer like polyethylene might precipitate out and clog injector orifices, valves, etc. Although, it is impossible to make a quantitative estimate concerning the relative dose above which this problem would become serious, polymerization may well represent the major radiation damage problem in ethylene.

Formation of Reactive Chemical Species

The formation of ozone in liquid oxygen has already been discussed. There have been other examples of this type of behavior, particularly in the case of the amine fuels. For example, small amounts of hydrazoic acid, HN_3, were observed to form in liquid ammonia[11] and evidence for some unidentified species was noted in a radiation study on hydrazine[13]. However, it is doubtful that this phenomenon could lead to serious problems in fuel storage systems because such reactive products ordinarily form at rates orders of magnitude lower than does non-condensable gas, for example.

RECOMMENDATIONS FOR FUTURE RESEARCH

Radiation Shielding Studies

It was concluded in the preceeding section that radiation emitted from α-heat sources would not be expected to seriously damage stored liquid propellants. However, parametric shielding studies may be desirable for larger α-heat sources to provide design data for space craft where other radiation sensitive components and/or instruments are present. These studies would be aimed at providing data for design of shielding with optimum weight and configuration. Such shielding studies would be necessary prior to spacecraft design if use of large ^{90}Sr, ^{60}Co, or reactor power sources were considered. Possible use of the fuel as shield material should be considered.

Radiation Research on Liquid Propellants

Research areas where definitive data are lacking have been mentioned in various sections of this report. Some of the areas where additional research would be desirable are the following:

(1) More complete radiation effects data are needed for boron hydride fuels, fluoride oxidizers, and liquid ethylene before one can estimate with reasonable certainty the storage life of these propellants in radiation fields.

(2) More definitive study of neutron effects on boron hydride fuels is needed to ascertain what problems may be raised by the ^{10}B$(n,\alpha)^{7}$Li reaction.

(3) It would be desirable to carry out long term storage tests of propellant systems at radiation dose rates more comparable to those expected on an actual spacecraft.

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